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(54)【発明の名称】 排気ガス浄化用触媒

(57)【要約】

【課題】 エンジン始動時の低温域において排出される炭化水素 (HC) を効率良く浄化する排気ガス浄化用触媒を提供すること。

【解決手段】 担体上に HC 吸着材層、耐熱性無機酸化物層及び触媒成分層を積層し、耐熱性無機酸化物層が γ -アルミナ、触媒成分層が Ce - [Y] - O_x 搅持 Pd や Ce - Al₂O₃ 搅持 Pd、を含んで成る排気ガス浄化用触媒である。担体上に積層した HC 吸着材層上且つ排気ガス流通方向に対し上流側に耐熱性無機酸化物層、下流側に触媒成分層を配設し、耐熱性無機酸化物層が γ -アルミナを含み、触媒成分層の第1層は Ce - [Y] - O_x 搅持 Pd や Ce - Al₂O₃ 搅持 Pd を含み、第2層は Ce - Al₂O₃ 搅持 Pt、Zr - Al₂O₃ 搅持 Rh 及び ZrO₂などを含む排気ガス浄化用触媒である。

【特許請求の範囲】

【請求項1】 担体上にH C吸着材層、耐熱性無機酸化物層及び触媒成分層を順次積層して成る排気ガス浄化用触媒であって、

上記耐熱性無機酸化物層が γ -アルミナを主成分として成り、上記触媒成分層がCe-[Y]-O_x担持Pd

(Yはジルコニウム、プラセオジム及びネオジムから成る群より選ばれた少なくとも1種の元素を示す)、並びに／又はCe-Al₂O₃担持Pdを主成分として成ることを特徴とする排気ガス浄化用触媒。

【請求項2】 担体上にH C吸着材層、耐熱性無機酸化物層及び触媒成分層を順次積層して成る排気ガス浄化用触媒であって、

上記耐熱性無機酸化物層が γ -アルミナを主成分として成り、上記触媒成分層が排気ガスの流通方向に対して上流側に配設される上流側触媒成分層と下流側に配設される下流側触媒成分層とから構成され、

該上流側触媒成分層はCe-[Y]-O_x担持Pd(Yはジルコニウム、プラセオジム及びネオジムから成る群より選ばれた少なくとも1種の元素を示す)、並びに／又はCe-Al₂O₃担持Pdを主成分として成り、該下流側触媒成分層はCe-Al₂O₃担持Pt、Zr-Al₂O₃担持Rh及びCe-ZrO₂から成る群より選ばれた少なくとも1種のものを主成分として成ることを特徴とする排気ガス浄化用触媒。

【請求項3】 担体上にH C吸着材層、耐熱性無機酸化物層及び触媒成分層を順次積層して成る排気ガス浄化用触媒であって、

上記耐熱性無機酸化物層が γ -アルミナを主成分として成り、上記触媒成分層が第1層及び第2層をこの順に積層して成り、

該第1層はCe-[Y]-O_x担持Pd(Yはジルコニウム、プラセオジム及びネオジムから成る群より選ばれた少なくとも1種の元素を示す)、並びに／又はCe-Al₂O₃担持Pdを主成分として成り、該第2層はCe-Al₂O₃担持Pt、Zr-Al₂O₃担持Rh及びZr含有CeO₂から成る群より選ばれた少なくとも1種のものを主成分として成ることを特徴とする排気ガス浄化用触媒。

【請求項4】 担体上にH C吸着材層を積層し、このH C吸着材層上且つ排気ガスの流通方向に対して上流側に耐熱性無機酸化物層、下流側に触媒成分層を配設して成る排気ガス浄化用触媒であって、

上記耐熱性無機酸化物層が γ -アルミナを主成分として成り、上記触媒成分層が第1層及び第2層をこの順に積層して成り、

該第1層はCe-[Y]-O_x担持Pd(Yはジルコニウム、プラセオジム及びネオジムから成る群より選ばれた少なくとも1種の元素を示す)、並びに／又はCe-Al₂O₃担持Pdを主成分として成り、該第2層はC

e-Al₂O₃担持Pt、Zr-Al₂O₃担持Rh及びZr含有CeO₂から成る群より選ばれた少なくとも1種のものを主成分として成ることを特徴とする排気ガス浄化用触媒。

【請求項5】 担体上且つ排気ガスの流通方向に対して、上流側にH C吸着材層、耐熱性無機酸化物層及び上流側触媒成分層をこの順に積層し、下流側に耐熱性無機酸化物層及び下流側触媒成分層をこの順に積層して成る排気ガス浄化用触媒であって、

10 上記耐熱性無機酸化物層が γ -アルミナを主成分として成り、上記上流側触媒成分層がCe-[Y]-O_x担持Pd(Yはジルコニウム、プラセオジム及びネオジムから成る群より選ばれた少なくとも1種の元素を示す)、並びに／又はCe-Al₂O₃担持Pdを主成分として成り、上記下流側触媒成分層がCe-Al₂O₃担持Pdを主成分として成ることを特徴とする排気ガス浄化用触媒。

【発明の詳細な説明】

【0001】

20 【発明の属する技術分野】本発明は、自動車(ガソリン、ディーゼル)、ボイラーなどの内燃機関から排出される排気ガス中の炭化水素(HC)、一酸化炭素(CO)及び窒素酸化物(NOx)を浄化する排気ガス浄化用触媒に係り、特に三元触媒が活性化しないエンジン始動時の低温域において大量に排出されるHCを浄化する排気ガス浄化用触媒に関する。

【0002】

【従来の技術】近年、内燃機関のエンジン始動時の低温域で大量に排出されるHC(コールドHC)の浄化を目的に、HC吸着材にゼオライトを用いたHC吸着型三元触媒(HC吸着機能付き三元触媒)が開発されている。

30 【0003】吸着材から脱離するHCの浄化触媒としては、従来ロジウム(Rh)、白金(Pt)及びパラジウム(Pd)等の貴金属種を同一層に共存させた仕様や、Rh層とPd層を塗り分けた仕様等が提案されている。例えば、特開平2-56247号公報には、ゼオライトを主成分とする第一層の上に、Pt、Pd及びRh等の貴金属を主成分とする第二層を設けた排気ガス浄化用触媒が提案されている。

40 【0004】また、かかるHC吸着材を用いた発明は、例えば、特開平6-74019号公報、特開平7-144119号公報、特開平6-142457号公報、特開平5-59942号公報及び特開平7-102957号公報等に開示されている。

【0005】特開平6-74019号公報には、排気流路にバイパスを設け、エンジン始動直後のコールド時に排出されるHCをバイパス流路に配置したHC吸着材に一旦吸着させ、その後流路を切換え、下流の三元触媒が活性化した後、排気ガスの一部をHC吸着触媒に通じ、脱離したHCを徐々に後段の三元触媒で浄化するシテ

ムが提案されている。また、特開平7-144119号公報には、コールド時に前段の三元触媒に熱を奪わせ中段のH C吸着触媒の吸着効率を向上し、前段の三元触媒活性化後は、タンデム配置した中段のH C吸着触材を介して後段の三元触媒に反応熱を伝熱し易くし、後段の三元触媒の浄化を促進するシステムが提案されている。更に、特開平6-1421457号公報には、低温域で吸着したH Cが脱離する際に、脱離H Cを含む排気ガスを熱交換器で予熱し三元触媒での浄化を促進するコールドH C吸着除去システムが提案されている。

【0006】一方、特開平5-59942号公報には、触媒配置とバルブによる排気ガスの流路を切換えによって、H C吸着材の昇温を緩慢にし、コールドH Cの吸着効率を向上するシステムが提案されている。また、特開平7-102957号公報には、後段の酸化・三元触媒の浄化性能を向上するため、前段の三元触媒と中段のH C吸着材の間に空気を供給し、後段の酸化・三元触媒の活性化を促進するシステムが提案されている。更に、特開平7-96183号公報には、ゼオライトと接触する触媒層(PdとAl₂O₃が主成分)の高温下での劣化抑制の観点から、吸着材層と触媒層との間に多孔質バリア層を設け、しかも、下層吸着材層の吸着能の低下を抑制するため、Pd担持Al₂O₃粒子の平均粒子径が15~25μm、耐火性無機質粒子の平均粒子径が5~15μmのものを用いることが提案されている。更にまた、特開平11-210451号公報には、排気ガス流れ方向に対して、H C吸着層と三元触媒層との配置を変更した構造によって、H C吸着層から脱離するH Cの浄化を促進することが提案されている。

【0007】

【発明が解決しようとする課題】吸着材としてゼオライトを用いる場合、コールドH C吸着性能は、排ガス中のH C種組成とゼオライトの有する細孔径との間に相関があるので、最適な細孔径と分布、骨格構造をもつゼオライトを使用することを要する。従来は、MFⅠ型をメインに、他の細孔径を有するゼオライト(例えば、USYなど)を単独で又はこれらを混合して、細孔径分布を調整していたが、耐久後にはゼオライト種によって細孔径の歪みや吸着・脱離特性が異なるため、排ガスH C種の吸着が不十分であるという問題点があった。

【0008】また、従来の炭化水素吸着材層と浄化触媒層を設けた排気ガス浄化用触媒では、内燃機関の始動直後の排気ガス低温域において炭化水素吸着材に吸着したコールドH Cが、排気ガス温度の上昇する前に脱離し浄化できない、更に、吸着H Cが脱離する際に、浄化触媒層の雰囲気は酸素不足状態になるため、理論空燃比域での浄化に有効な三元触媒では、H C、CO及びNO_xのバランスの良い浄化ができなくなり、吸着H Cの浄化が十分でないという問題点があった。この対応策として、排気流路の切換えによって、三元触媒が十分に活性化し

た後、吸着H Cを脱離させ三元触媒で浄化する方法、電熱ヒーターによって三元触媒の早期活性化を図る方法、及び外部から空気を導入して三元触媒の活性化開始を速める方法なども検討されているが、システム構成が煩雑化ししかも十分なコールドH Cの低減効果が得られない、コストが高い、長期間の使用に耐えられない、などの問題点があった。

【0009】更に、炭化水素吸着材と浄化触媒と組合せ逐次積層した一体構造型触媒においても、内燃機関の始動直後の排気ガス低温域において炭化水素吸着材に吸着したコールドH Cが、排気ガス温度の上昇する前に脱離し浄化できない、更に、吸着H Cの脱離と三元触媒の活性化域とが重なった場合においても、十分に活性化していない三元触媒では、H C、CO及びNO_xのバランスの良い浄化ができなくなり、吸着H Cの浄化が十分にできないという問題点があった。

【0010】本発明は、このような従来技術の有する課題に鑑みてなされたものであり、その目的とするところは、エンジン始動時の低温域において排出される炭化水素(H C)を効率良く浄化する排気ガス浄化用触媒を提供することにある。

【0011】

【課題を解決するための手段】本発明者は上記課題を解決すべく鋭意研究を重ねた結果、耐熱性無機酸化物層により、H C吸着材層で吸着・保持した炭化水素(H C)の脱離速度、脱離量を抑制することにより、上記課題が解決できることを見出し、本発明を完成するに至った。

【0012】即ち、本発明の排気ガス浄化用触媒は、担体上にH C吸着材層、耐熱性無機酸化物層及び触媒成分層を順次積層して成る排気ガス浄化用触媒であって、上記耐熱性無機酸化物層がγ-アルミナを主成分として成り、上記触媒成分層がCe-[Y]-O_x担持Pd(Yはジルコニウム、プラセオジム及びネオジムから成る群より選ばれた少なくとも1種の元素を示す)、並びに/又はCe-Al₂O₃担持Pdを主成分として成ることを特徴とする。

【0013】また、本発明の排気ガス浄化用触媒は、担体上にH C吸着材層、耐熱性無機酸化物層及び触媒成分層を順次積層して成る排気ガス浄化用触媒であって、上記耐熱性無機酸化物層がγ-アルミナを主成分として成り、上記触媒成分層が排気ガスの流通方向に対して上流側に配設される上流側触媒成分層と下流側に配設される下流側触媒成分層とから構成され、該上流側触媒成分層はCe-[Y]-O_x担持Pd(Yはジルコニウム、プラセオジム及びネオジムから成る群より選ばれた少なくとも1種の元素を示す)、並びに/又はCe-Al₂O₃担持Pdを主成分として成り、該下流側触媒成分層はCe-Al₂O₃担持Pt、Zr-Al₂O₃担持Rh及びCe-ZrO₂から成る群より選ばれた少なくとも1種のものを主成分として成ることを特徴とする。

【0014】更に、本発明の排気ガス浄化用触媒は、担体上にHC吸着材層、耐熱性無機酸化物層及び触媒成分層を順次積層して成る排気ガス浄化用触媒であって、上記耐熱性無機酸化物層が γ -アルミナを主成分として成り、上記触媒成分層が第1層及び第2層をこの順に積層して成り、該第1層はCe-[Y]-O_x担持Pd(Yはジルコニウム、プラセオジム及びネオジムから成る群より選ばれた少なくとも1種の元素を示す)、並びに/又はCe-Al₂O₃担持Pdを主成分として成り、該第2層はCe-Al₂O₃担持Pt、Zr-Al₂O₃担持Rh及びZr含有CeO₂から成る群より選ばれた少なくとも1種のものを主成分として成ることを特徴とする。

【0015】更にまた、本発明の排気ガス浄化用触媒は、担体上にHC吸着材層を積層し、このHC吸着材層上且つ排気ガスの流通方向に対して上流側に耐熱性無機酸化物層、下流側に触媒成分層を配設して成る排気ガス浄化用触媒であって、上記耐熱性無機酸化物層が γ -アルミナを主成分として成り、上記触媒成分層が第1層及び第2層をこの順に積層して成り、該第1層はCe-[Y]-O_x担持Pd(Yはジルコニウム、プラセオジム及びネオジムから成る群より選ばれた少なくとも1種の元素を示す)、並びに/又はCe-Al₂O₃担持Pdを主成分として成り、該第2層はCe-Al₂O₃担持Pt、Zr-Al₂O₃担持Rh及びZr含有CeO₂から成る群より選ばれた少なくとも1種のものを主成分として成ることを特徴とする。

【0016】また、本発明の排気ガス浄化用触媒は、担体上且つ排気ガスの流通方向に対して、上流側にHC吸着材層、耐熱性無機酸化物層及び上流側触媒成分層をこの順に積層し、下流側に耐熱性無機酸化物層及び下流側触媒成分層をこの順に積層して成る排気ガス浄化用触媒であって、上記耐熱性無機酸化物層が γ -アルミナを主成分として成り、上記上流側触媒成分層がCe-[Y]-O_x担持Pd(Yはジルコニウム、プラセオジム及びネオジムから成る群より選ばれた少なくとも1種の元素を示す)、並びに/又はCe-Al₂O₃担持Pdを主成分として成り、上記下流側触媒成分層がCe-Al₂O₃担持Pdを主成分として成ることを特徴とする。

【0017】

【発明の実施の形態】以下、本発明の排気ガス浄化用触媒について詳細に説明する。なお、本明細書において「%」は、特記しない限り質量百分率を示す。また、図2~13に、本発明の一例及び比較例の触媒構造を示すが、これらの図では排気ガスが左側から右側へ流通することを想定している。

【0018】本発明の排気ガス浄化用触媒は、HC吸着材層が吸着した排気ガス中のHCを、触媒成分層が放出する酸素と効率良く反応させることにより、HC、CO及びNO_x、特に三元触媒が活性化しないエンジン始動

時の低温域のHCを効率良く浄化する。具体的には、耐熱性無機酸化物層を設けて、HC吸着材層のHC脱離を遅延化し、また、触媒成分層に含まれる触媒成分が低温・酸素不足雰囲気で優れた酸素放出能や触媒活性を発現することにより、脱離HCが効率良く浄化される。本発明者は、かかる効果を発現させる排気ガス浄化用触媒として、以下の5つの触媒構造を見出した。

【0019】まず、本発明の第1の排気ガス浄化用触媒は、担体上にHC吸着材層、耐熱性無機酸化物層及び触媒成分層を順次積層して成る。ここで、上記耐熱性無機酸化物層は γ -アルミナを主成分として成り、上記触媒成分層はCe-[Y]-O_x担持Pd(Yはジルコニウム(Zr)、プラセオジム(Pr)又はネオジム(Nd)、及びこれらの任意の組合せに係る元素を示す)、並びに/又はCe-Al₂O₃担持Pdを主成分として成る。例えば、図3に示す触媒構造を挙げることができる。この場合は、吸着HCの拡散方向上又は排気ガスの流通方向上に設けられた耐熱性無機酸化物層により、HC吸着材層に拡散しそれぞれ内を通過する排気ガスの流通量又は流通速度が減少するため、吸着HCの脱離が遅延化され、脱離HCを効率良く浄化することができる。また、上記耐熱性無機酸化物層として用いる γ -アルミナは、平均粒子径が1~3μmであることが緻密である(ゼオライトに比べてガス透過性が低い)面から望ましい。この上限以下の方がより吸着HCの脱離遅延化効果が大きく、この下限以上の方がより吸着HCの拡散が早くなり吸着効率の低下が起こりにくい。

【0020】また、本発明の第2の排気ガス浄化用触媒は、触媒成分層が上流側触媒成分層及び下流側触媒成分層から成ること以外は、上記第1の触媒とほぼ同様の構成を有する。即ち、担体上にHC吸着材層、耐熱性無機酸化物層及び触媒成分層を順次積層して成り、この触媒成分層が、排気ガスの流通方向に対して上流側に配設される上流側触媒成分層と下流側に配設される下流側触媒成分層とから構成される。ここで、上流側触媒成分層はCe-[Y]-O_x担持Pd(YはZr、Pr又はNd、及びこれらの任意の組合せに係る元素を示す)、並びに/又はCe-Al₂O₃担持Pdを主成分として成り、下流側触媒成分層はCe-Al₂O₃担持Pt、Zr-Al₂O₃担持Rh又はCe-ZrO₂、及びこれらの任意の組合せに係るものを主成分として成る。例えば、図5に示すような触媒構造を挙げることができる。この場合は、HC吸着材の温度上昇が緩慢になり、また吸着HCの脱離が遅延化されるので有効である。また、上述の上流側触媒成分層及び下流側触媒成分層を使用することで、下流に酸素不足の雰囲気でのHC浄化能にすぐれる触媒成分層を配置することができ、脱離HCの浄化効率を更に向上することができる。

【0021】更に、本発明の第3の排気ガス浄化用触媒は、触媒成分層を2層構造としたこと以外は、上記第1

の触媒とほぼ同様の構成を有する。即ち、担体上にH C吸着材層、耐熱性無機酸化物層及び触媒成分層を順次積層して成り、上記触媒成分層が第1層及び第2層をこの順に積層して成る。ここで、上記第1層はCe-[Y]-O_x担持Pd (YはZr、Pr又はNd、及びこれらの任意の組合せに係る元素を示す)、並びに/又はCe-Al₂O₃担持Pdを主成分として成り、上記第2層はCe-Al₂O₃担持Pt、Zr-Al₂O₃担持Rh又はZr含有CeO₂ (ZC20)、及びこれらの任意の組合せに係るものを主成分として成る。例えば、図6及び図7に示すような触媒構造を挙げることができる。

【0022】次に、本発明の第4の排気ガス浄化用触媒は、担体上にH C吸着材層を積層し、このH C吸着材層上且つ排気ガスの流通方向に対して上流側に耐熱性無機酸化物層、下流側に触媒成分層を配設して成る。ここで、上記耐熱性無機酸化物層はγ-アルミナを主成分として成り、上記触媒成分層は第1層及び第2層をこの順に積層して成る。また、上記第1層はCe-[Y]-O_x担持Pd (YはZr、Pr又はNd、及びこれらの任意の組合せに係る元素を示す)、並びに/又はCe-Al₂O₃担持Pdを主成分として成り、上記第2層はCe-Al₂O₃担持Pt、Zr-Al₂O₃担持Rh又はZr含有CeO₂、及びこれらの任意の組合せに係るものを主成分として成る。例えば、図2に示すような触媒構造を挙げができる。この場合は、上流側の温度上昇を抑え、しかも吸着H Cの脱離遅延化が図られるため下流での脱離H C浄化効率が向上する。

【0023】次に、本発明の第5の排気ガス浄化用触媒は、担体上且つ排気ガスの流通方向に対して、上流側にH C吸着材層、耐熱性無機酸化物層及び上流側触媒成分層をこの順に積層し、下流側に耐熱性無機酸化物層及び下流側触媒成分層をこの順に積層して成る。ここで、上記耐熱性無機酸化物層はγ-アルミナを主成分として成り、上記上流側触媒成分層はCe-[Y]-O_x担持Pd (YはZr、Pr又はNd、及びこれらの任意の組合せに係る元素を示す)、並びに/又はCe-Al₂O₃担持Pdを主成分として成り、上記下流側触媒成分層がCe-Al₂O₃担持Pdを主成分として成る。例えば、図4に示すような触媒構造を挙げができる。この場合は、下流側に設けた耐熱性無機酸化物層により、H CがH C吸着材層内に拡散しそれぞれ構造内を通過する排気ガス量が減少する。また、吸着H Cは脱離が抑制されるとともに耐熱性無機酸化物層上に設けた下流側净化触媒成分層で効率良く浄化される。なお、耐熱性無機酸化物層の塗布量を調整して排気ガスと上流側触媒成分層及び下流側净化触媒成分層との接触状態を良好にすることことができ、このときはH C吸着層から脱離する吸着H Cと上流側触媒成分層から放出された酸素とを、下流側净化触媒成分層で効率良く浄化することができ

る。

【0024】なお、上述した排気ガス浄化用触媒のH C吸着材層の構成成分としては、代表的に、β-ゼオライト、モルデナイト、USY及びMFIなどを使用することができます。これらを用いるといろいろな分子径のH C種を効率良く吸着できるので有効である。また、上記排気ガス浄化用触媒では、排気ガスの流通方向に対して上流側に下流側より多くの酸素放出材及び貴金属を含有することができる。これより、上流側における酸素放出量を下流側より増大させることができるので、脱離された吸着H Cを効率良く浄化できる。

【0025】更に、上記担体としては、代表的に一体構造型ハニカム担体を例示できる。また、担体における各成分の塗布面積は適宜調整することが望ましく、例えば、図2の排気ガス浄化用触媒では、担体の上流側から下流側へ且つ全長の5~50%の範囲に亘って耐熱性無機酸化物層が配設されていることが望ましい。更に、図4の排気ガス浄化用触媒では、担体の上流側から下流側へ且つ全長の20~60%の範囲に亘って上流側触媒成分層が配設されていることが望ましい。更にまた、図5の排気ガス浄化用触媒では、担体の上流側から下流側へ且つ全長の50~90%の範囲に亘って上流側净化触媒成分層が配設されていることが望ましい。これらの場合は、脱離H Cと放出酸素との反応効率を向上できるので有効である。

【0026】

【実施例】以下、本発明を実施例及び比較例により更に詳細に説明するが、本発明はこれら実施例に限定されるものではない。

30 【0027】(実施例1)

・H C吸着材層

Si/2Al比が3.5のβ-ゼオライト粉末800g、シリカゾルを1333.3g (固体分濃度15%)と純水1000gをアルミナ製ボールミルポットに投入し、60分間粉碎してスラリー液を得た。このスラリー液をモノリス担体 (300セル/6ミル、触媒容量1.0L)に付着させ、空気流にてセル内の余剰のスラリーを取り除いて、50℃の空気流通下30分間乾燥し、次いで、150℃の空気流通下15分間乾燥した後、400℃で1時間焼成した。この時の塗布量として、焼成後に350g/Lになるまでコーティング作業を繰り返し、触媒-aを得た。

【0028】・耐熱性無機酸化物層

γ-アルミナ950gと硝酸酸性アルミナゾル500g、純水1000gを磁性ボールミルに投入し、混合粉碎してスラリー液を得た。このスラリー液を上記触媒-aの排気ガス上流側の1/4に付着させ、空気流にてセル内の余剰のスラリーを取り除いて乾燥し、400℃で1時間焼成し、コート層重量35g/Lを塗布し、触媒-aを得た。

【0029】・触媒成分層(第1層)

Ce 3 mol %を含むアルミナ粉末(A 197 mol %)に、硝酸パラジウム水溶液を含浸或いは高速攪拌中で噴霧し、150℃で24時間乾燥した後、400℃で1時間、次いで、600℃で1時間焼成し、Pd担持アルミナ粉末(粉末a)を得た。この粉末aのPd濃度は4.0%であった。La 1 mol %とZr 3.2 mol %含有セリウム酸化物粉末(Ce 6.7 mol %)に、硝酸パラジウム水溶液を含浸或いは高速攪拌中で噴霧し、150℃で24時間乾燥した後、400℃で1時間、次いで、600℃で1時間焼成し、Pd担持セリウム酸化物粉末(粉末b)を得た。この粉末bのPd濃度は2.0%であった。上記Pd担持アルミナ粉末(粉末a)40.0 g、Pd担持セリウム酸化物粉末(粉末b)14.1 g、硝酸酸性アルミナゾル24.0 g(ペーマイトアルミナ10%に10%の硝酸を添加することによって得られたゾルでA12O3換算で24 g)及び炭酸バリウム100 g(BaOとして6.7 g)を純水2000 gを磁性ボールミルに投入し、混合粉碎してスラリー液を得た。このスラリー液を上記コート触媒-a 1の排気ガス下流側の3/4に付着させ、空気流にてセル内の余剰のスラリーを取り除いて乾燥し、400℃で1時間焼成し、コート層重量66.5 g/Lを塗布し、触媒-bを得た。

【0030】・触媒成分層(第2層)

Zr 3 mol %を含むアルミナ粉末(A 197 mol %)に、硝酸ロジウム水溶液を含浸或いは高速攪拌中で噴霧し、150℃で24時間乾燥した後、400℃で1時間、次いで、600℃で1時間焼成し、Rh担持アルミナ粉末(粉末c)を得た。この粉末cのRh濃度は2.0%であった。Ce 3 mol %を含むアルミナ粉末(A 197 mol %)に、ジニトロジアンミン白金水溶液を含浸或いは高速攪拌中で噴霧し、150℃で24時間乾燥した後、400℃で1時間、次いで、600℃で1時間焼成し、Pt担持アルミナ粉末(粉末d)を得た。この粉末dのPt濃度は3.0%であった。La 1 モル%とCe 2.0 モル%を含有するジルコニア酸化物粉末に、ジニトロジアンミン白金水溶液を含浸或いは高速攪拌中で噴霧し、150℃で24時間乾燥した後、400℃で1時間、次いで、600℃で1時間焼成し、Pt担持アルミナ粉末(粉末e)を得た。この粉末dのPt濃度は3.0%であった。上記Rh担持アルミナ粉末(粉末c)11.8 g、Pt担持アルミナ粉末(粉末d)11.8 g、Pt担持ジルコニア酸化物粉末(粉末e)11.8 g、硝酸酸性アルミナゾル16.0 gを磁性ボールミルに投入し、混合粉碎してスラリー液を得た。このスラリー液を上記コート触媒触媒-a 1の排気ガス下流側の3/4に付着させ、空気流にてセル内の余剰のスラリーを取り除いて乾燥し、400℃で1時間焼成し、コート層重量37 g/Lを塗布し、触媒を得た(図2)。触媒の貴金属担持量は、Pt 0.71 g/L、Pd 1.8

8 g/L、Rh 0.24 g/Lであった。

【0031】(実施例2) γ-アルミナ950 gと硝酸酸性アルミナゾル500 g、純水1000 gを磁性ボールミルに投入し、混合粉碎してスラリー液を得た。このスラリー液を上記触媒-aの排気ガス上流側の1/4に付着させ、空気流にてセル内の余剰のスラリーを取り除いて乾燥し、400℃で1時間焼成し、コート層重量35 g/Lを塗布し、触媒-a 2を得た。実施例1と同様の操作を繰り返して、触媒-a 2に三元触媒成分層を塗布し、図3に示す構造を有する本例の触媒を得た。

【0032】(実施例3~6) Zr 3 mol %を含むアルミナ粉末(A 197 mol %)に、硝酸ロジウム水溶液を含浸或いは高速攪拌中で噴霧し、150℃で24時間乾燥した後、400℃で1時間、次いで、600℃で1時間焼成し、Rh担持アルミナ粉末(粉末e)を得た。この粉末eのRh濃度は1.5%であった。Ce 3 mol %を含むアルミナ粉末(A 197 mol %)に、ジニトロジアンミン白金水溶液を含浸或いは高速攪拌中で噴霧し、150℃で24時間乾燥した後、400℃で1時間、次いで、600℃で1時間焼成し、Pt担持アルミナ粉末(粉末f)を得た。この粉末dのPt濃度は1.5%であった。実施例1又は実施例2とほぼ同様の操作を繰り返して、図4~7に示す構造を有する実施例3~6の触媒を得た。

【0033】(三元触媒(TWC))粉末aを53.0 g、粉末bを23.6 gと硝酸酸性アルミナゾル7.0 g(ペーマイトアルミナ10%に10%の硝酸を添加することによって得られたゾルでA_{1.2}O₃換算で14 g)及び炭酸バリウム4.0 g(BaOとして2.7 g)を純水1000 gを磁性ボールミルに投入し、混合粉碎してスラリー液を得た。このスラリー液をモノリス担体(900セル/2ミル、触媒容量1.0 L)に付着させ、空気流にてセル内の余剰のスラリーを取り除いて乾燥し、400℃で1時間焼成し、コート層重量7.8 g/Lを塗布し、触媒-fを得た。

【0034】粉末eを31.3 g、La 1 モル%とCe 2.0 モル%を含有するジルコニア酸化物粉末を100 gと硝酸酸性アルミナゾル17.0 g(ペーマイトアルミナ10%に10%の硝酸を添加することによって得られたゾルでA_{1.2}O₃換算で17 g)を純水1000 gを磁性ボールミルに投入し、混合粉碎してスラリー液を得た。このスラリー液を触媒-fに付着させ、空気流にてセル内の余剰のスラリーを取り除いて乾燥し、400℃で1時間焼成し、コート層重量4.3 g/Lを塗布し、触媒を得た。触媒の貴金属担持量は、Pd 2.35 g/L、Rh 0.47 g/Lであった。

【0035】(比較例1) γ-アルミナを主成分とする緻密な耐火性無機酸化物層を有しない構造とした以外は、実施例1と同様の操作を繰り返して、図8に示す構造を有する本例の触媒を得た。

【0036】(比較例2) γ -アルミナを主成分とする緻密な耐火性無機酸化物層を有しない構造とした以外は、実施例2と同様の操作を繰り返して、図9示す構造を有する本例の触媒を得た。

【0037】(比較例3) γ -アルミナを主成分とする緻密な耐火性無機酸化物層を有しない構造とした以外は、実施例3と同様の操作を繰り返して、図10に示す構造を有する本例の触媒を得た。

【0038】(比較例4) γ -アルミナを主成分とする緻密な耐火性無機酸化物材層を有しない構造とした以外は、実施例4と同様の操作を繰り返して、図11に示す構造を有する本例の触媒を得た。

【0039】(比較例5) γ -アルミナを主成分とする緻密な耐火性無機酸化物材層を有しない構造とした以外は、実施例5と同様の操作を繰り返して、図12に示す構造を有する本例の触媒を得た。

【0040】(比較例6) γ -アルミナを主成分とする緻密な耐火性無機酸化物材層を有しない構造とした以外は、実施例6と同様の操作を繰り返して、図13に示す*

*構造を有する本例の触媒を得た。

【0041】実施例1～6及び比較例1～6で得られた触媒仕様を表1に示す。また、実施例7～12及び比較例7～12として、図1に示すように、上流側に三元触媒(TWC)を配設した評価装置を用いて評価試験を行った。この結果を表2に示す。

【0042】<評価方法>

・耐久条件

10	エンジン排気量	3000cc
	燃料	ガソリン(日石ダッシュ)
	触媒入口ガス温度	650℃
	耐久時間	100時間
	・車両性能試験	
	エンジン排気量	日産自動車株式会社製 直列4気筒 2.0Lエンジン
	評価方法	北米排ガス試験法のLA4-CHの A-bag

【0043】

【表1】

	Pt (g/L)	Pd (g/L)	Rh (g/L)	BaO (g/L)	La0.01Ce0.89[X]0.3Ox [X]
実施例1	0.71	1.88	0.24	6.7	Zr
実施例2	—	2.83	—	3.4	Pr
実施例3	—	2.83	—	3.4	Nd
実施例4	0.71	1.88	0.24	6.7	Pr0.2Nd0.1
実施例5	0.71	1.88	0.24	6.7	Zr0.2Pr0.1
実施例6	0.71	1.88	0.24	6.7	Zr
TWC	—	2.36	0.47	3.4	Zr
比較例1	0.71	1.88	0.24	6.7	Zr
比較例2	—	2.83	—	3.4	Pr
比較例3	—	2.83	—	3.4	Nd
比較例4	0.71	1.88	0.24	6.7	Pr0.2Nd0.1
比較例5	0.71	1.88	0.24	6.7	Zr0.2Pr0.1
比較例6	0.71	1.88	0.24	6.7	Zr

【0044】

【表2】

	触媒位置1	触媒位置1	吸着率(%)	脱離浄化率(%)
実施例7	TWC	実施例1の触媒	79	37
実施例8	TWC	実施例2の触媒	78	38
実施例9	TWC	実施例3の触媒	78	39
実施例10	TWC	実施例4の触媒	79	39
実施例11	TWC	実施例5の触媒	79	38
実施例12	TWC	実施例6の触媒	78	37
比較例7	TWC	比較例1の触媒	78	32
比較例8	TWC	比較例2の触媒	79	33
比較例9	TWC	比較例3の触媒	78	34
比較例10	TWC	比較例4の触媒	79	34
比較例11	TWC	比較例5の触媒	78	33
比較例12	TWC	比較例6の触媒	79	32

【0045】表2より、実施例1～6で得られた触媒は、比較例1～6で得られた触媒よりもHC脱離浄化能が優れていることがわかる。

【0046】以上、本発明を実施例により詳細に説明したが、本発明はこれらに限定されるものではなく、本発明の要旨の範囲内において種々の変形が可能である。例えば、排気ガス流路を変形させることや各触媒成分の担持量を変化させることにより、排気ガスの接触率や接触時間を調整することができる。

【0047】

【発明の効果】以上説明してきたように、本発明によれば、耐熱性無機酸化物層により、HC吸着材層で吸着・保持した炭化水素(HC)の脱離速度、脱離量を抑制することとしたため、エンジン始動時の低温域において排出される炭化水素(HC)を効率良く浄化する排気ガス*

*浄化用触媒を提供することができる。

【図面の簡単な説明】

【図1】評価装置の構造を示す概略図である。

【図2】実施例1の触媒構造を示す概略図である。

【図3】実施例2の触媒構造を示す概略図である。

【図4】実施例3の触媒構造を示す概略図である。

【図5】実施例4の触媒構造を示す概略図である。

【図6】実施例5の触媒構造を示す概略図である。

【図7】実施例6の触媒構造を示す概略図である。

【図8】比較例1の触媒構造を示す概略図である。

【図9】比較例2の触媒構造を示す概略図である。

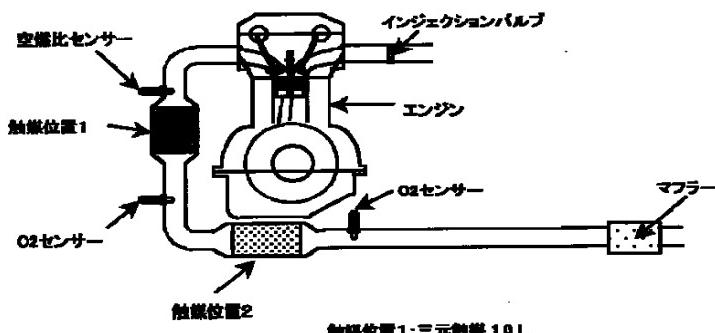
【図10】比較例3の触媒構造を示す概略図である。

【図11】比較例4の触媒構造を示す概略図である。

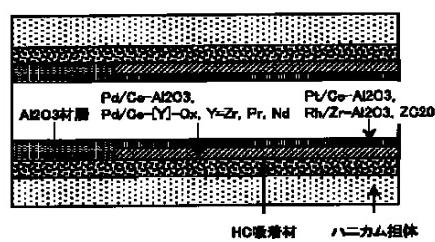
【図12】比較例5の触媒構造を示す概略図である。

【図13】比較例6の触媒構造を示す概略図である。

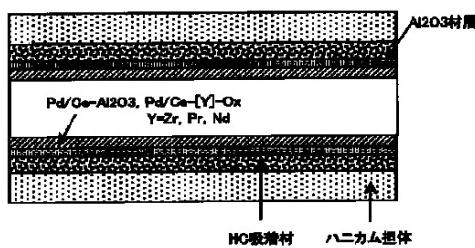
【図1】



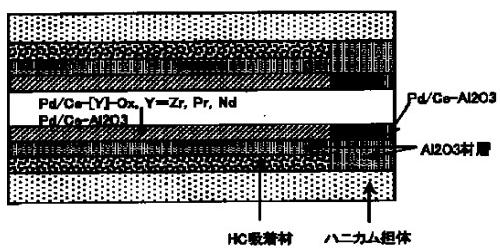
【図2】



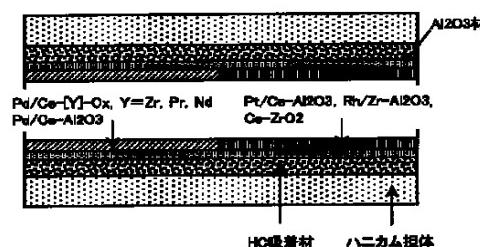
【図3】



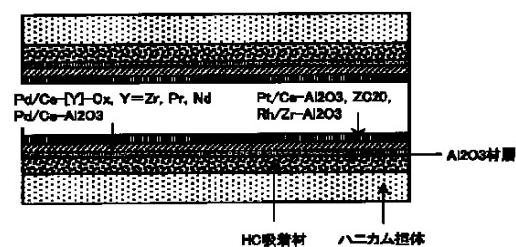
【図4】



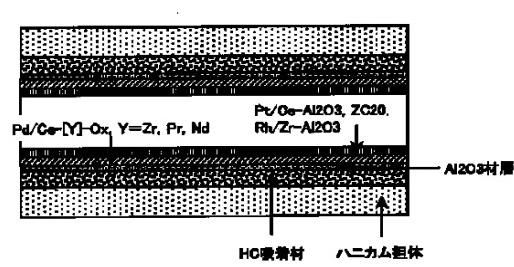
【図5】



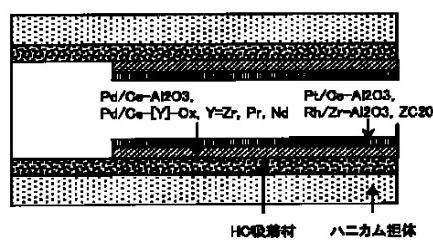
【図6】



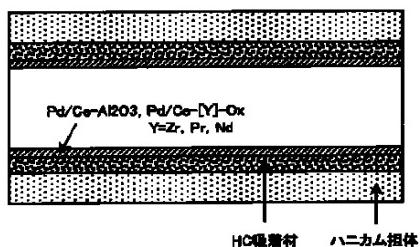
【図7】



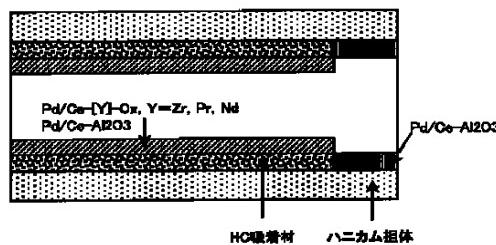
【図8】



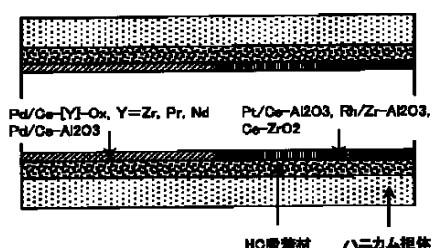
【図9】



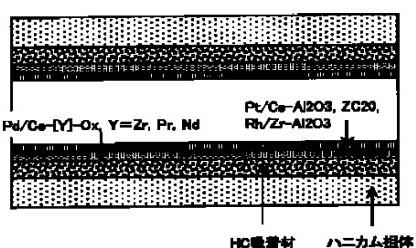
【図10】



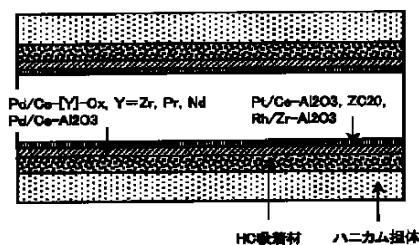
【図11】



【図13】



【図12】



フロントページの続き

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			Z A B

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GB07W GB09Y GB10W HA20
HA36 HA37 HA42
4D048 AA06 AA13 AA18 AB05 BA03X
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CA09 EA19 EB12Y EC28
EC29 ZA19A ZA19B

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Notes:

1. Untranslatable words are replaced with asterisks (****).
2. Texts in the figures are not translated and shown as it is.

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Dictionary: Last updated 12/14/2009 / Priority: 1. Chemistry / 2. Mechanical engineering

CLAIM + DETAILED DESCRIPTION

[Claim(s)]

[Claim 1]It is a catalyst for exhaust gas purification which laminates an HC adsorbent layer, a heat-resistant inorganic oxide layer, and a catalyst component layer one by one on a carrier, the above-mentioned heat-resistant inorganic oxide layer changes considering gamma-alumina as a principal component -- the above-mentioned catalyst component layer -- the Ce-[Y]-Ox support Pd (Y -- zirconium.) A catalyst for exhaust gas purification changing considering/or the Ce-aluminum₂O₃ support Pd as a principal component and at least one sort of elements chosen from a group which comprises praseodymium and neodymium are shown.

[Claim 2]It is a catalyst for exhaust gas purification which laminates an HC adsorbent layer, a heat-resistant inorganic oxide layer, and a catalyst component layer one by one on a carrier, The above-mentioned heat-resistant inorganic oxide layer changes considering gamma-alumina as a principal component, and it comprises an upstream catalyst component layer by which the above-mentioned catalyst component layer is allocated in the upstream to a circulation direction of exhaust gas, and a downstream catalyst component layer allocated in the downstream, this upstream catalyst component layer -- the Ce-[Y]-Ox support Pd (Y -- zirconium.) . At least one sort of elements chosen from a group which comprises praseodymium and neodymium are shown. And it changes considering/or the Ce-aluminum₂O₃ support Pd as a principal component, A catalyst for exhaust gas purification, wherein this downstream catalyst component layer changes considering at least one sort of things chosen from a group which comprises the Ce-aluminum₂O₃ support Pt, the Zr-aluminum₂O₃ support Rh, and Ce-ZrO₂ as a principal component.

[Claim 3]It is a catalyst for exhaust gas purification which laminates an HC adsorbent layer, a heat-resistant inorganic oxide layer, and a catalyst component layer one by one on a carrier, The above-mentioned heat-resistant inorganic oxide layer changes considering gamma-

alumina as a principal component, and the above-mentioned catalyst component layer laminates the 1st layer and the 2nd layer in this order. This 1st layer The Ce-[Y]-Ox support Pd (Y shows at least one sort of elements chosen from a group which comprises zirconium, praseodymium, and neodymium), And it changes considering/or the Ce-aluminum₂O₃ support Pd as a principal component, A catalyst for exhaust gas purification, wherein this 2nd layer changes considering at least one sort of things chosen from a group which comprises the Ce-aluminum₂O₃ support Pt, the Zr-aluminum₂O₃ support Rh, and Zr content CeO₂ as a principal component.

[Claim 4]Laminate an HC adsorbent layer on a carrier and to the upstream to this HC adsorbent layer top and a circulation direction of exhaust gas A heat-resistant inorganic oxide layer, Are a catalyst for exhaust gas purification which allocates a catalyst component layer in the downstream, and the above-mentioned heat-resistant inorganic oxide layer changes considering gamma-alumina as a principal component, the above-mentioned catalyst component layer laminates the 1st layer and the 2nd layer in this order -- this -- the 1st layer -- the Ce-[Y]-Ox support Pd (Y -- zirconium.) . At least one sort of elements chosen from a group which comprises praseodymium and neodymium are shown. And it changes considering/or the Ce-aluminum₂O₃ support Pd as a principal component, A catalyst for exhaust gas purification, wherein this 2nd layer changes considering at least one sort of things chosen from a group which comprises the Ce-aluminum₂O₃ support Pt, the Zr-aluminum₂O₃ support Rh, and Zr content CeO₂ as a principal component.

[Claim 5]An HC adsorbent layer, a heat-resistant inorganic oxide layer, and an upstream catalyst component layer are laminated to the upstream to a carrier top and a circulation direction of exhaust gas at this order, It is a catalyst for exhaust gas purification which laminates a heat-resistant inorganic oxide layer and a downstream catalyst component layer to the downstream at this order, the above-mentioned heat-resistant inorganic oxide layer changes considering gamma-alumina as a principal component -- the above-mentioned upstream catalyst component layer -- the Ce-[Y]-Ox support Pd (Y -- zirconium.) . At least one sort of elements chosen from a group which comprises praseodymium and neodymium are shown. And a catalyst for exhaust gas purification, wherein it changes considering/or the Ce-aluminum₂O₃ support Pd as a principal component and the above-mentioned downstream catalyst component layer changes considering the Ce-aluminum₂O₃ support Pd as a principal component.

[Detailed Description of the Invention]

[0001]

[Field of the Invention]The hydrocarbon (HC) in the exhaust gas in which this invention is discharged from motors, such as a car (gasoline, diesel) and a boiler, It is related with the catalyst for exhaust gas purification which purifies HC discharged in large quantities in the low temperature region at the time of the engine start which starts the catalyst for exhaust gas purification which purifies carbon monoxide (CO) and nitrogen oxides (NOx), especially a three way component catalyst does not activate.

[0002]

[Description of the Prior Art]In recent years, HC adsorption type three way component catalyst (three way component catalyst with HC adsorbing function) which used zeolite for HC adsorbent is developed for the purpose of purification of HC (cold HC) discharged in large quantities in the low temperature region at the time of the engine start of a motor.

[0003]As a purifying catalyst of HC desorbed from adsorption material, the specification which made noble-metals kinds, such as rhodium (Rh), platinum (Pt), and palladium (Pd), live together in the same layer conventionally, the specification which distinguished Rh layer and Pd layer by different color with, etc. are proposed. For example, the catalyst for exhaust gas purification which provided the secondary layer which uses noble metals, such as Pt, Pd, and Rh, as a principal component on the first layer that uses zeolite as a principal component is proposed by JP,H2-56247,A.

[0004]The invention using this HC adsorbent is indicated by JP,H6-74019,A, JP,H7-144119,A, JP,H6-142457,A, JP,H5-59942,A, JP,H7-102957,A, etc., for example.

[0005]Form a bypass in an exhaust passage at JP,H6-74019,A, and HC discharged at the time of cold one immediately after engine start is made to once stick to the HC adsorbent arranged to the bypass passage, After switching a pass after that and activating a downstream three way component catalyst, the system which purifies gradually HC desorbed through the HC adsorbing catalyst from some exhaust gas with a latter three way component catalyst is proposed. Make JP,H7-144119,A take heat to the three way component catalyst of the preceding paragraph at the time of cold one, improve the adsorption efficiency of the HC adsorbing catalyst of the middle, and, [after three way component catalyst activation of the preceding paragraph] The system which makes the heat transmission of the heat of reaction easy to carry out to a latter three way component catalyst via HC ***** material of the middle which carried out tandem arrangement, and promotes purification of a latter three way component catalyst is proposed. When HC to which it stuck in the low temperature region ****s, cold HC adsorption treatment system which preheats exhaust gas including the desorption HC with a heat exchanger, and promotes purification with a three way component catalyst is proposed by JP,H6-1421457,A.

[0006]On the other hand, temperature up of HC adsorbent is made slow for the pass of the

exhaust gas by catalyst arrangement and a valve by change, and the system which improves the adsorption efficiency of cold HC is proposed by JP,H5-59942,A. In order to improve the purification performance of latter oxidation and three way component catalyst, air is supplied between the three way component catalyst of the preceding paragraph, and the HC adsorbent of the middle, and the system which promotes activation of latter oxidation and three way component catalyst is proposed by JP,H7-102957,A. . [JP,H7-96183,A] [from a viewpoint of the deterioration inhibition under the elevated temperature of the catalyst bed (Pd and aluminum₂O₃ are principal components) in contact with zeolite] In order to provide a porosity barrier layer between an adsorption material layer and a catalyst bed and to control the fall of the adsorption capacity of a lower layer adsorption material layer moreover, it is proposed that the mean particle diameter of Pd support aluminum₂O₃ grains uses that whose mean particle diameter of 15-25 micrometers and a fireproof inorganic particle is 5-15 micrometers.

Promoting the purification of HC desorbed from a hydrocarbon adsorption layer to the direction of an exhaust gas flow by the structure which changed arrangement with a hydrocarbon adsorption layer and a three way component catalyst layer is proposed by JP,H11-210451,A again.

[0007]

[Problem to be solved by the invention] Since cold HC adsorption performance has correlation between HC kind presentation in exhaust gas, and the pole diameter which zeolite has when using zeolite as adsorption material, it requires using zeolite with the optimal pole diameter and distribution, and skeletal structure. Although independent or these were mixed for the zeolite (for example, USY etc.) which has other pole diameters for a MFI type in a main and pore volume distribution was adjusted conventionally, After durability, since distortion and adsorption / desorption characteristics of a pole diameter changed with zeolite kinds, the problem that adsorption of an exhaust gas HC kind was insufficient was.

[0008][a catalyst for exhaust gas purification which provided a conventional hydrocarbon adsorption material layer and a purifying catalyst layer] Since atmosphere of a purifying catalyst layer will be in an insufficient oxygen state when [in which cold HC which stuck to hydrocarbon adsorption material in an exhaust gas low temperature region immediately after start up of a motor cannot **** and purify before exhaust gas temperature rises] the adsorption HC ****'s further, There was a problem that good purification of balance of HC, CO, and NOx became impossible, and a three way component catalyst effective in purification in a theoretical-air-fuel-ratio region was not enough as purification of the adsorption HC. After a three way component catalyst is fully activated by the change of an exhaust passage as this measure, Although a method of desorbing the adsorption HC and purifying with a three way component catalyst, a method of attaining early activation of a three way component catalyst with an electrical heater, a method of introducing air from the exterior and speeding up an

activation start of a three way component catalyst, etc. are examined, There were problems -- high-cost prolonged use from which system structure becomes complicated and a reduction effect of sufficient cold HC moreover is not acquired cannot be borne.

[0009]Also in an integral-construction type catalyst which was combined with hydrocarbon adsorption material and a purifying catalyst, and was laminated one by one, Also in a case which cold HC which stuck to hydrocarbon adsorption material in an exhaust gas low temperature region immediately after start up of a motor cannot **** and purify before exhaust gas temperature rises where desorption of the adsorption HC and an activation region of a three way component catalyst lap further, In a three way component catalyst which is not fully being activated, there was a problem that good purification of balance of HC, CO, and NOx became impossible, and purification of the adsorption HC could not fully be performed.

[0010]this invention is made in view of SUBJECT which such conventional technology has, and comes out. The purpose is to provide the catalyst for exhaust gas purification which purifies efficiently the hydrocarbon (HC) discharged in the low temperature region at the time.

[0011]

[Means for solving problem]As a result of repeating research wholeheartedly that above-mentioned SUBJECT should be solved, by controlling the desorption rate of the hydrocarbon (HC) adsorbed and held in the HC adsorbent layer, and a desorption amount by a heat-resistant inorganic oxide layer, this invention person finds out that above-mentioned SUBJECT is solvable, and came to complete this invention.

[0012]Namely, the catalyst for exhaust gas purification of this invention is a catalyst for exhaust gas purification which laminates an HC adsorbent layer, a heat-resistant inorganic oxide layer, and a catalyst component layer one by one on a carrier, the above-mentioned heat-resistant inorganic oxide layer changes considering gamma-alumina as a principal component -- the above-mentioned catalyst component layer -- the Ce-[Y]-Ox support Pd (Y -- zirconium.) And at least one sort of elements chosen from the group which comprises praseodymium and neodymium are shown, it changes considering/or the Ce-aluminum₂O₃ support Pd as a principal component.

[0013]The catalyst for exhaust gas purification of this invention is a catalyst for exhaust gas purification which laminates an HC adsorbent layer, a heat-resistant inorganic oxide layer, and a catalyst component layer one by one on a carrier, The above-mentioned heat-resistant inorganic oxide layer changes considering gamma-alumina as a principal component, and it comprises an upstream catalyst component layer by which the above-mentioned catalyst component layer is allocated in the upstream to the circulation direction of exhaust gas, and a downstream catalyst component layer allocated in the downstream, this upstream catalyst component layer -- the Ce-[Y]-Ox support Pd (Y -- zirconium.) . At least one sort of elements

chosen from the group which comprises praseodymium and neodymium are shown. And it changes considering/or the Ce-aluminum₂O₃ support Pd as a principal component, This downstream catalyst component layer changes considering at least one sort of things chosen from the group which comprises the Ce-aluminum₂O₃ support Pt, the Zr-aluminum₂O₃ support Rh, and Ce-ZrO₂ as a principal component.

[0014]A catalyst for exhaust gas purification of this invention is a catalyst for exhaust gas purification which laminates an HC adsorbent layer, a heat-resistant inorganic oxide layer, and a catalyst component layer one by one on a carrier, The above-mentioned heat-resistant inorganic oxide layer changes considering gamma-alumina as a principal component, and the above-mentioned catalyst component layer laminates the 1st layer and the 2nd layer in this order. This 1st layer The Ce-[Y]-Ox support Pd (Y shows at least one sort of elements chosen from a group which comprises zirconium, praseodymium, and neodymium), And it changes considering/or the Ce-aluminum₂O₃ support Pd as a principal component, This 2nd layer changes considering at least one sort of things chosen from a group which comprises the Ce-aluminum₂O₃ support Pt, the Zr-aluminum₂O₃ support Rh, and Zr content CeO₂ as a principal component.

[0015]A catalyst for exhaust gas purification of this invention laminates an HC adsorbent layer on a carrier again, To this HC adsorbent layer top and a circulation direction of exhaust gas, to the upstream A heat-resistant inorganic oxide layer, Are a catalyst for exhaust gas purification which allocates a catalyst component layer in the downstream, and the above-mentioned heat-resistant inorganic oxide layer changes considering gamma-alumina as a principal component, the above-mentioned catalyst component layer laminates the 1st layer and the 2nd layer in this order -- this -- the 1st layer -- the Ce-[Y]-Ox support Pd (Y -- zirconium.) . At least one sort of elements chosen from a group which comprises praseodymium and neodymium are shown. And it changes considering/or the Ce-aluminum₂O₃ support Pd as a principal component, This 2nd layer changes considering at least one sort of things chosen from a group which comprises the Ce-aluminum₂O₃ support Pt, the Zr-aluminum₂O₃ support Rh, and Zr content CeO₂ as a principal component.

[0016]A catalyst for exhaust gas purification of this invention receives a carrier top and a circulation direction of exhaust gas, An HC adsorbent layer, a heat-resistant inorganic oxide layer, and an upstream catalyst component layer are laminated to the upstream at this order, It is a catalyst for exhaust gas purification which laminates a heat-resistant inorganic oxide layer and a downstream catalyst component layer to the downstream at this order, the above-mentioned heat-resistant inorganic oxide layer changes considering gamma-alumina as a principal component -- the above-mentioned upstream catalyst component layer -- the Ce-[Y]-

Ox support Pd (Y -- zirconium.) And at least one sort of elements chosen from a group which comprises praseodymium and neodymium are shown, it changes considering/or the Ce-aluminum₂O₃ support Pd as a principal component, and the above-mentioned downstream catalyst component layer changes considering the Ce-aluminum₂O₃ support Pd as a principal component.

[0017]

[Mode for carrying out the invention]Hereafter, a catalyst for exhaust gas purification of this invention is explained in detail. Unless it mentions specially "%" in this Description, percentage by mass is shown. Although catalyst structure of an example of this invention and a comparative example is shown in drawing 2 - 13, in these figures, it assumes that exhaust gas circulates from left-hand side to right-hand side.

[0018]A catalyst for exhaust gas purification of this invention purifies efficiently HC of a low temperature region at the time of engine start which HC, CO and NOx, especially a three way component catalyst do not activate by making HC in exhaust gas to which an HC adsorbent layer stuck react to oxygen which a catalyst component layer emits efficiently. When a catalyst component which provides a heat-resistant inorganic oxide layer, and specifically delay-izes HC desorption of an HC adsorbent layer, and is contained in a catalyst component layer reveals oxygen discharge ability and catalytic activity excellent in low temperature and insufficient oxygen atmosphere, the desorption HC is purified efficiently. this invention person found out the following five catalyst structures as a catalyst for exhaust gas purification which makes this effect reveal.

[0019]First, the 1st catalyst for exhaust gas purification of this invention laminates an HC adsorbent layer, a heat-resistant inorganic oxide layer, and a catalyst component layer one by one on a carrier. The above-mentioned heat-resistant inorganic oxide layer changes considering gamma-alumina as a principal component here, The above-mentioned catalyst component layer changes considering the Ce-[Y]-Ox support Pd (Y shows zirconium (Zr), praseodymium (Pr) or neodymium (Nd), and an element concerning these arbitrary combination) and/, or the Ce-aluminum₂O₃ support Pd as a principal component. For example, catalyst structure shown in drawing 3 can be mentioned. In this case, in order that turn volume or velocity of circulation of exhaust gas which is spread in an HC adsorbent layer and passes through inside of a zeolite layer by a heat-resistant inorganic oxide layer provided on a diffusion direction of the adsorption HC or a circulation direction of exhaust gas may decrease, Desorption of the adsorption HC is delay-ized and the desorption HC can be purified efficiently. As for gamma-alumina used as the above-mentioned heat-resistant inorganic oxide layer, it is desirable from a precise field (compared with zeolite, gas permeability is low) for mean particle diameter to be 1-3 micrometers. A desorption delay-ized effect of the adsorption HC is [below

this maximum] larger, and, in diffusion of the adsorption HC, decline in adsorption efficiency does not take place [a direction more than this lower limit] more easily early.

[0020]The 2nd catalyst for exhaust gas purification of this invention has the almost same composition as the 1st catalyst of the above except a catalyst component layer comprising an upstream catalyst component layer and a downstream catalyst component layer. That is, an HC adsorbent layer, a heat-resistant inorganic oxide layer, and a catalyst component layer are laminated one by one on a carrier, and this catalyst component layer comprises an upstream catalyst component layer allocated in the upstream to a circulation direction of exhaust gas, and a downstream catalyst component layer allocated in the downstream. Here, an upstream catalyst component layer is the Ce-[Y]-Ox support Pd (). [Y] [Zr and] And Pr or Nd, and an element concerning these arbitrary combination are shown, it changes considering/or the Ce-aluminum₂O₃ support Pd as a principal component, A downstream catalyst component layer changes considering the Ce-aluminum₂O₃ support Pt, the Zr-aluminum₂O₃ support Rh or Ce-ZrO₂, and a thing concerning these arbitrary combination as a principal component. For example, catalyst structure as shown in drawing 5 can be mentioned. In this case, since a rise in heat of HC adsorbent becomes slow and desorption of the adsorption HC is delay-ized, it is effective. By using an above-mentioned upstream catalyst component layer and a downstream catalyst component layer, a catalyst component layer which is excellent in HC decontamination capacity in atmosphere of insufficient oxygen can be arranged downstream, and purification efficiency of the desorption HC can be improved further.

[0021]The 3rd catalyst for exhaust gas purification of this invention has the almost same composition as the 1st catalyst of the above except having made the catalyst component layer into two-layer structure. That is, an HC adsorbent layer, a heat-resistant inorganic oxide layer, and a catalyst component layer are laminated one by one on a carrier, and the above-mentioned catalyst component layer laminates the 1st layer and the 2nd layer in this order. here -- the 1st above-mentioned layer -- the Ce-[Y]-Ox support Pd (Y -- Zr and Pr -- or) [Nd and] And and the element concerning these arbitrary combination is shown, it changes considering/or the Ce-aluminum₂O₃ support Pd as a principal component, The 2nd above-mentioned layer changes considering the Ce-aluminum₂O₃ support Pt, the Zr-aluminum₂O₃ support Rh or Zr content CeO₂ (ZC20), and the thing concerning these arbitrary combination as a principal component. For example, catalyst structure as shown in drawing 6 and drawing 7 can be mentioned.

[0022]Next, the 4th catalyst for exhaust gas purification of this invention laminates an HC adsorbent layer on a carrier, and to this HC adsorbent layer top and the circulation direction of exhaust gas, a heat-resistant inorganic oxide layer is allocated in the upstream, and it allocates a catalyst component layer in the downstream. Here, the above-mentioned heat-resistant

inorganic oxide layer changes considering gamma-alumina as a principal component, and the above-mentioned catalyst component layer laminates the 1st layer and the 2nd layer in this order. the 1st above-mentioned layer -- the Ce-[Y]-Ox support Pd (Y -- Zr and Pr -- or) [Nd and] And and the element concerning these arbitrary combination is shown, it changes considering/or the Ce-aluminum₂O₃ support Pd as a principal component, The 2nd above-mentioned layer changes considering the Ce-aluminum₂O₃ support Pt, the Zr-aluminum₂O₃ support Rh or Zr content CeO₂, and the thing concerning these arbitrary combination as a principal component. For example, catalyst structure as shown in drawing 2 can be mentioned. In this case, the rise in heat of the upstream is suppressed, and since desorption delay-ization of the adsorption HC can moreover be attained, the desorption HC purifying efficiency in the lower stream improves.

[0023]Next, to a carrier top and the circulation direction of exhaust gas, the 5th catalyst for exhaust gas purification of this invention laminates an HC adsorbent layer, a heat-resistant inorganic oxide layer, and an upstream catalyst component layer to the upstream at this order, and laminates a heat-resistant inorganic oxide layer and a downstream catalyst component layer to the downstream at this order. The above-mentioned heat-resistant inorganic oxide layer changes considering gamma-alumina as a principal component here, the above-mentioned upstream catalyst component layer -- the Ce-[Y]-Ox support Pd (Y -- Zr and Pr -- or) [Nd and] And and the element concerning these arbitrary combination is shown, it changes considering/or the Ce-aluminum₂O₃ support Pd as a principal component, and the above-mentioned downstream catalyst component layer changes considering the Ce-aluminum₂O₃ support Pd as a principal component. For example, catalyst structure as shown in drawing 4 can be mentioned. In this case, the exhaust gas volume which HC is spread in an HC adsorbent layer and passes through the inside of zeolite structure by the heat-resistant inorganic oxide layer provided in the downstream decreases. The adsorption HC is efficiently purified in the downstream purifying catalyst component layer provided on the heat-resistant inorganic oxide layer while desorption is controlled. The coverage of a heat-resistant inorganic oxide layer can be adjusted, and a contact state with exhaust gas, an upstream catalyst component layer, and a downstream purifying catalyst component layer can be made good, The adsorption HC desorbed from a hydrocarbon adsorption layer at this time and the oxygen emitted from the upstream catalyst component layer can be efficiently purified in a downstream purifying catalyst component layer.

[0024]As a composition component of an HC adsorbent layer of a catalyst for exhaust gas purification mentioned above, beta-zeolite, mordenite, USY, MFI, etc. can be used typically. Since HC kind of various molecular diameters can be efficiently adsorbed if these are used, it is effective. In the above-mentioned catalyst for exhaust gas purification, many oxygen

discharge material and noble metals can be contained from the downstream in the upstream to a circulation direction of exhaust gas. From this, since an oxygen burst size in the upstream can be increased from the downstream, the adsorption HC from which it was desorbed can be purified efficiently.

[0025]As the above-mentioned carrier, an integral-construction type honeycomb carrier can be illustrated typically. moreover -- as for a coated area of each component in a carrier, adjusting suitably is desirable -- a catalyst for exhaust gas purification of drawing 2 -- the downstream from the upstream of a carrier -- and it is desirable to allocate a heat-resistant inorganic oxide layer [5 to 50% of range of an overall length]. furthermore -- a catalyst for exhaust gas purification of drawing 4 -- the downstream from the upstream of a carrier -- and it is desirable to allocate an upstream catalyst component layer [20 to 60% of range of an overall length]. furthermore -- again -- a catalyst for exhaust gas purification of drawing 5 -- the downstream from the upstream of a carrier -- and it is desirable to allocate an upstream purifying catalyst component layer [50 to 90% of range of an overall length]. Since reaction efficiency of the desorption HC and discharge oxygen can be improved in these cases, it is effective.

[0026]

[Working example]Hereafter, although an embodiment and a comparative example explain this invention still in detail, this invention is not limited to these embodiments.

[0027](Embodiment 1)

- HC adsorbent layer Si / 2aluminum ratio -- the beta-zeolite powder 800g of 35, and silica -- 1333.3 g (15% of solids concentration) and the pure water 1000g were fed into a ball mill pot made from alumina, sol was ground for 60 minutes, and slurry liquid was obtained. After made this slurry liquid adhere to a monolith carrier (300 cells /6mil, catalyst capacity 1.0L), having removed slurry of a surplus in a cell in an air style, having dried for 30 minutes under 50 ** air circulation, ranking second and drying for 15 minutes under 150 ** air circulation, it calcinated at 400 ** for 1 hour. As coverage at this time, coating work was repeated until it became 350 g/L after calcination, and catalyst-a was obtained.

[0028]- 950 g of heat-resistant inorganic oxide layer gamma-alumina, and 500 g of nitric acid acidity alumina sol and the pure water 1000g were fed into a magnetic ball mill, preferential grinding was carried out, and slurry liquid was obtained. This slurry liquid was made to adhere to one fourth of the exhaust gas upstream of the above-mentioned catalyst-a, and in an air style, slurry of a surplus in a cell was removed, and it dried, and calcinated at 400 ** for 1 hour, coated layer weight of 35 g/L was applied, and catalyst-a1 was obtained.

[0029]- A catalyst component layer (the 1st layer)

After a palladium nitrate aqueous solution's having high-speed been under being impregnated or churning to alumina powder (aluminum97mol%) containing Ce3mol%, spraying on it and drying at 150 ** to it for 24 hours, it ranked second at 400 ** for 1 hour, and calcinated at 600

** for 1 hour, and Pd support alumina powder (powder a) was obtained. Pd concentration of this powder a was 4.0%. After a palladium nitrate aqueous solution's high-speed being under being impregnated or churning to La1mol% and Zr32mol% content cerium oxide powder (Ce67mol%), spraying on them and drying at 150 ** to them for 24 hours, it ranks second at 400 ** for 1 hour, It calcinated at 600 ** for 1 hour, and Pd support cerium oxide powder (powder b) was obtained. Pd concentration of this powder b was 2.0%. 400 g of the above-mentioned Pd support alumina powder (powder a), Pd support cerium oxide powder (powder b) 141, 240g (.) of nitric acid acidity alumina sol [by adding 10% of nitric acid to boehmite alumina 10%] By obtained sol, the pure water 2000g was fed into a magnetic ball mill, preferential grinding of 24g and the barium carbonate 100g (67g as BaO) was carried out by Al₂O₃ conversion, and slurry liquid was obtained. This slurry liquid was made to adhere to three fourths of the exhaust gas downstream of above-mentioned coat catalyst-a1, and in an air style, slurry of a surplus in a cell was removed, and it dried, and calcinated at 400 ** for 1 hour, coated layer weight of 66.5 g/L was applied, and catalyst-b was obtained.

[0030]- A catalyst component layer (the 2nd layer)

After a rhodium nitrate aqueous solution's having high-speed been under being impregnated or churning to alumina powder (aluminum97mol%) containing Zr3mol%, spraying on it and drying at 150 ** to it for 24 hours, it ranked second at 400 ** for 1 hour, and calcinated at 600 ** for 1 hour, and Rh support alumina powder (powder c) was obtained. Rh concentration of this powder c was 2.0%. After a dinitrodiammine platinum solution's having high-speed been under being impregnated or churning to alumina powder (aluminum97mol%) containing Ce3mol%, spraying on it and drying at 150 ** to it for 24 hours, it ranked second at 400 ** for 1 hour, and calcinated at 600 ** for 1 hour, and Pt support alumina powder (powder d) was obtained. Pt concentration of this powder d was 3.0%. After a dinitrodiammine platinum solution's having high-speed been under being impregnated or churning to zirconium oxide powder containing La1 mol % and Ce20 mol %, spraying on it and drying at 150 ** to it for 24 hours, it ranked second at 400 ** for 1 hour, and calcinated at 600 ** for 1 hour, and Pt support alumina powder (powder e) was obtained. Pt concentration of this powder d was 3.0%. 118 g of the above-mentioned Rh support alumina powder (powder c), 118 g of Pt support alumina powder (powder d), 118 g of Pt support zirconium oxide powder (powder e), and 160 g of nitric acid acidity alumina sol were supplied to a magnetic ball mill, preferential grinding was carried out, and slurry liquid was obtained. This slurry liquid was made to adhere to three fourths of the exhaust gas downstream of above-mentioned coat catalyst catalyst-a1, and in an air style, slurry of a surplus in a cell was removed, and it dried, and calcinated at 400 ** for 1 hour, coated layer weight of 37 g/L was applied, and a catalyst was acquired (drawing 2). Noble-metals holding amounts of a catalyst were Pt0.71 g/L, Pd1.88 g/L, and Rh0.24 g/L.

[0031](Embodiment 2) 950 g of gamma-alumina, and 500 g of nitric acid acidity alumina sol

and pure water 1000g ** were supplied to a magnetic ball mill, preferential grinding was carried out, and slurry liquid was obtained. This slurry liquid was made to adhere to one fourth of the exhaust gas upstream of the above-mentioned catalyst-a, and in an air style, slurry of a surplus in a cell was removed, and it dried, and calcinated at 400 ** for 1 hour, coated layer weight of 35 g/L was applied, and catalyst-a2 was obtained. The same operation as Embodiment 1 was repeated, a three way component catalyst component layer was applied to catalyst-a2, and a catalyst of this example which has the structure shown in drawing 3 was acquired.

[0032](Embodiments 3-6) After a rhodium nitrate aqueous solution's having high-speed been under being impregnated or churning to alumina powder (aluminum97mol%) containing Zr3mol%, spraying on it and drying at 150 ** to it for 24 hours, it ranked second at 400 ** for 1 hour, and calcinated at 600 ** for 1 hour, and Rh support alumina powder (powder e) was obtained. Rh concentration of this powder e was 1.5%. After a dinitrodiammine platinum solution's having high-speed been under being impregnated or churning to alumina powder (aluminum97mol%) containing Ce3mol%, spraying on it and drying at 150 ** to it for 24 hours, it ranked second at 400 ** for 1 hour, and calcinated at 600 ** for 1 hour, and Pt support alumina powder (powder f) was obtained. Pt concentration of this powder d was 1.5%. The almost same operation as Embodiment 1 or Embodiment 2 was repeated, and a catalyst of Embodiments 3-6 which have the structure shown in drawing 4 - 7 was acquired.

[0033](Three way component catalyst (TWC)) The powder a 530 g, They are 236 g and 70g (.) of nitric acid acidity alumina sol about the powder b. [by adding 10% of nitric acid to boehmite alumina 10%] By the obtained sol, the pure water 1000g was fed into the magnetic ball mill, preferential grinding of 14g and the barium carbonate 40g (27g as BaO) was carried out by aluminum₂O₃ conversion, and slurry liquid was obtained. This slurry liquid was made to adhere to a monolith carrier (900 cells /2mil, catalyst capacity 1.0L), and in the air style, slurry of the surplus in a cell was removed, and it dried, and calcinated at 400 ** for 1 hour, the coated layer weight of 78 g/L was applied, and catalyst-f was obtained.

[0034]The powder e 313 g, They are 100 g and 170g (.) of nitric acid acidity alumina sol about the zirconium oxide powder containing La1 mol % and Ce20 mol %. [by adding 10% of nitric acid to boehmite alumina 10%] By the obtained sol, the pure water 1000g was fed into the magnetic ball mill, preferential grinding of the 17 g was carried out by aluminum₂O₃ conversion, and slurry liquid was obtained. This slurry liquid was made to adhere to catalyst-f, and in the air style, slurry of the surplus in a cell was removed, and it dried, and calcinated at 400 ** for 1 hour, the coated layer weight of 43 g/L was applied, and the catalyst was acquired. The noble-metals holding amounts of the catalyst were Pd2.35 g/L and Rh0.47 g/L.

[0035](Comparative example 1) Except having considered it as the structure where it does not have a precise refractory inorganic oxide layer which uses gamma-alumina as a principal

component, the same operation as Embodiment 1 was repeated, and the catalyst of this example which has the structure shown in drawing 8 was acquired.

[0036](Comparative example 2) Except having considered it as the structure where it does not have a precise refractory inorganic oxide layer which uses gamma-alumina as a principal component, the same operation as Embodiment 2 was repeated, and the catalyst of this example which has the structure shown figure 9 was acquired.

[0037](Comparative example 3) Except having considered it as the structure where it does not have a precise refractory inorganic oxide layer which uses gamma-alumina as a principal component, the same operation as Embodiment 3 was repeated, and the catalyst of this example which has the structure shown in drawing 10 was acquired.

[0038](Comparative example 4) Except having considered it as the structure where it does not have a precise refractory inorganic oxide material layer which uses gamma-alumina as a principal component, the same operation as Embodiment 4 was repeated, and the catalyst of this example which has the structure shown in drawing 11 was acquired.

[0039](Comparative example 5) Except having considered it as structure where it does not have a precise refractory inorganic oxide material layer which uses gamma-alumina as a principal component, the same operation as Embodiment 5 was repeated, and a catalyst of this example which has the structure shown in drawing 12 was acquired.

[0040](Comparative example 6) Except having considered it as structure where it does not have a precise refractory inorganic oxide material layer which uses gamma-alumina as a principal component, the same operation as Embodiment 6 was repeated, and a catalyst of this example which has the structure shown in drawing 13 was acquired.

[0041]Catalyst specification obtained by Embodiments 1-6 and the comparative examples 1-6 is shown in Table 1. As Embodiments 7-12 and the comparative examples 7-12, as shown in drawing 1, an evaluation test was done on the upstream using an evaluation system which allocated a three way component catalyst (TWC). This result is shown in Table 2.

[0042]A <valuation method> and an amount of durable condition engine exhaust gases 3000-cc fuel Gasoline (Nippon Oil dash)

Catalyst inlet gas temperature 650 ** endurance time 100 hour and, the amount of vehicle performance examination engine exhaust gases NISSAN MOTOR CO. LTD. make An in-series 4-cylinder 2.0L engine valuation method A-bag of LA4-CH of the North America exhaust gas examining method [0043]

[Table 1]

	Pt (g/L)	Pd (g/L)	Rh (g/L)	BaO (g/L)	La0.01Ce0.89[X]0.3Ox [X]
実施例1	0.71	1.88	0.24	6.7	Zr
実施例2	—	2.83	—	3.4	Pr
実施例3	—	2.83	—	3.4	Nd
実施例4	0.71	1.88	0.24	6.7	Pr0.2Nd0.1
実施例5	0.71	1.88	0.24	6.7	Zr0.2Pr0.1
実施例6	0.71	1.88	0.24	6.7	Zr
TWC	—	2.35	0.47	3.4	Zr
比較例1	0.71	1.88	0.24	6.7	Zr
比較例2	—	2.83	—	3.4	Pr
比較例3	—	2.83	—	3.4	Nd
比較例4	0.71	1.88	0.24	6.7	Pr0.2Nd0.1
比較例5	0.71	1.88	0.24	6.7	Zr0.2Pr0.1
比較例6	0.71	1.88	0.24	6.7	Zr

[0044]

[Table 2]

	触媒位置1	触媒位置1	吸着率(%)	脱離浄化率(%)
実施例7	TWC	実施例1の触媒	79	37
実施例8	TWC	実施例2の触媒	78	38
実施例9	TWC	実施例3の触媒	78	39
実施例10	TWC	実施例4の触媒	79	39
実施例11	TWC	実施例5の触媒	79	38
実施例12	TWC	実施例6の触媒	78	37
比較例7	TWC	比較例1の触媒	78	32
比較例8	TWC	比較例2の触媒	79	33
比較例9	TWC	比較例3の触媒	78	34
比較例10	TWC	比較例4の触媒	79	34
比較例11	TWC	比較例5の触媒	78	33
比較例12	TWC	比較例6の触媒	79	32

[0045]Table 2 shows that a catalyst acquired in Embodiments 1-6 excels a catalyst acquired by the comparative examples 1-6 in HC desorption decontamination capacity.

[0046]As mentioned above, although an embodiment explained this invention in detail, this

invention is not limited to these and various modification is possible for it within the limits of a gist of this invention. For example, a contact interval and contact time of exhaust gas can be adjusted by changing a holding amount of changing an exhaust gas passage or each catalyst component.

[0047]

[Effect of the Invention]It writes with controlling the desorption rate of the hydrocarbon (HC) adsorbed and held in the HC adsorbent layer, and a desorption amount by a heat-resistant inorganic oxide layer according to this invention, as explained above, The catalyst for exhaust gas purification which purifies efficiently the hydrocarbon (HC) discharged in the low temperature region at the time of engine start can be provided.

[Translation done.]